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The geochemical fate of Se(IV) in the Boom Clay system – XAS based solid phase speciation

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For more than 30 years the Boom Clay formation is studied as a reference host formation for methodological research concerning clay-based geological disposal of HLRW in Belgium and Europe. Boom Clay provides good sorption capacity, very low permeability and chemically reducing conditions due to the anoxic conditions and the presence of pyrite and siderite. Performance Assessment calculations have indicated Se^{79} ($t_{1/2} = 2.95\times10^5$ y) to be one of the critical radionuclides for the geological disposal of HLRW [1].

Aqueous selenite [Se(+IV)] and selenate [Se(+VI)] are the dominant species in mildly and strongly oxidizing environments. Under reducing conditions the solubility of Se is theoretically controlled by the formation of sparsely soluble selenium phases such as elemental Se or transition metal-selenide salts (e.g. FeSe or $FeSe_2$) [2, 3]. Slow kinetic reactions between the different redox states have been observed [4] and proposed to explain different redox phases observed within a single reducing environment. Se oxyanions, such as SeO_4^{2-} and SeO_3^{2-}, are generally considered as the most mobile forms of Se [5] and their migration through Boom Clay thus is considered as “worst case scenario”. In order to assess their long-term fate it is imperative to understand the influence of different geochemical phases present in the Boom Clay matrix on selenium speciation and mobility.

A multidisciplinary approach combining long-term batch sorption experiments with linear combination XANES and ITFA-based EXAFS analysis on different fractions isolated from Boom Clay batch systems equilibrated with Se(IV), identified Se^0 as the dominant in situ solid phase speciation of Se in Boom Clay conditions.

[1] SAFIR-2, 2001. Brussels, Belgium. p. 288. [2] Breynaert, E. *et al.*, ES&T. **42** (10): p. 3595. [3] Scheinost, A.C. *et al.*, J. Contam. Hydrol. **102** p. 228. [4] Masscheleyn, P.H. *et al.*, ES&T. **24** (1): p. 91. [5] Elrashidi, M.A. *et al.*, Soil Science. **144** (2): p. 141.